Transient XUV and X-ray lasers pumped by Free-Electron Laser Sources

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mpsd

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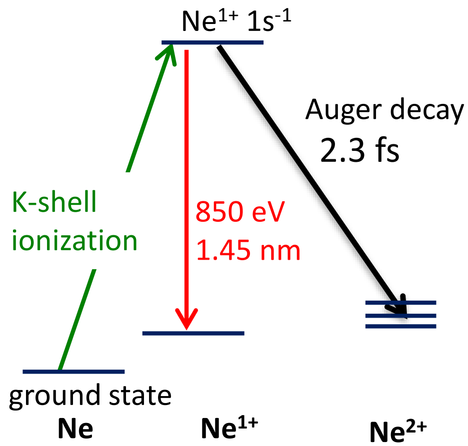
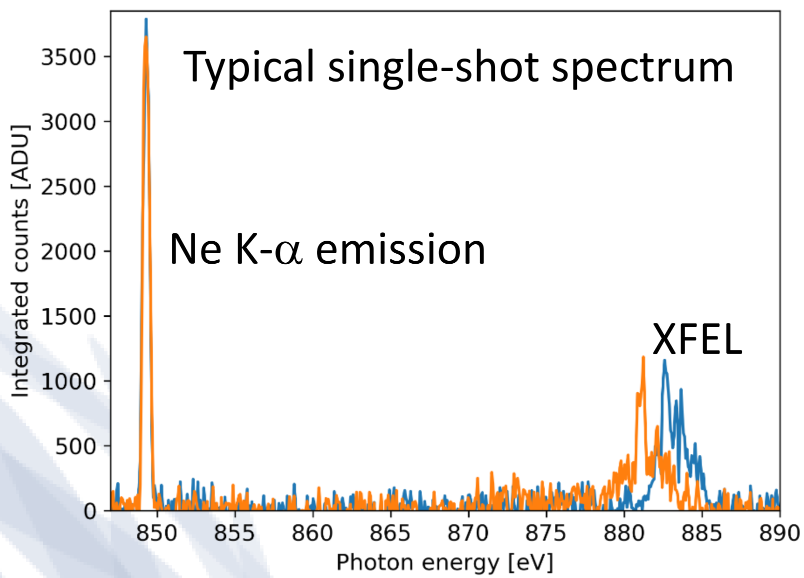
Amplified spontaneous x-ray emission

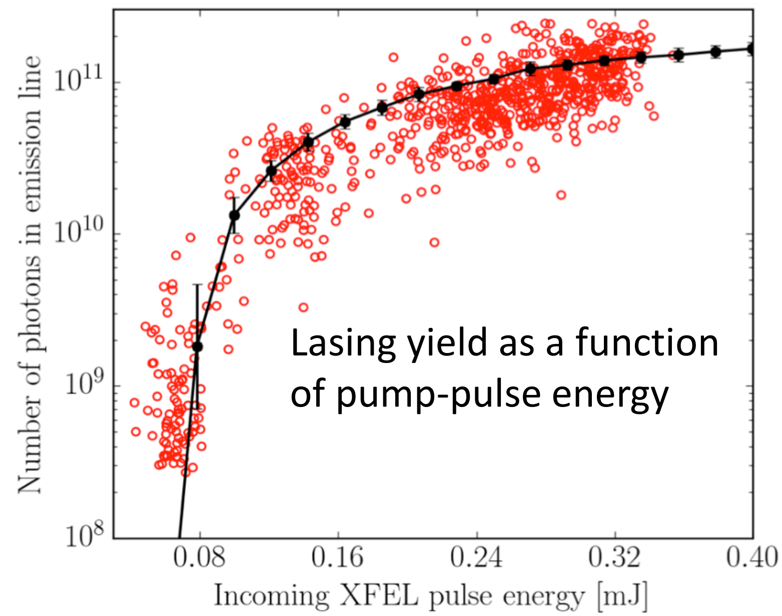
1st demonstration of photoionization Neon K-α laser at 1.45 nm



Photoionisation x-ray laser proposed by
Duguay and Rentzepis, Appl. Phys. Lett. 10, 350 (1967).

To overcome Auger-decay requires an x-ray pump with intensities $>10^{17}$ W/cm^2





- emission in forward direction
- amplification up to e^{21}
- transform limited x-ray pulses of fs duration
- Conversion efficiency reaching 10%

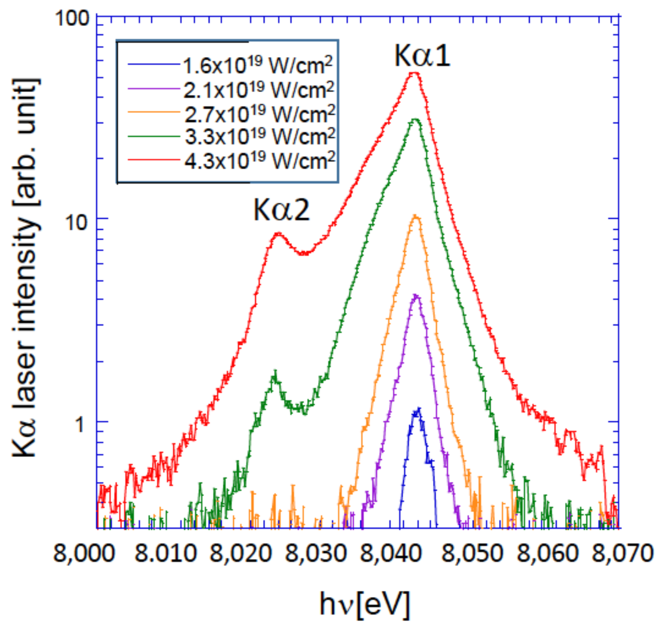
Rohringer et al., *Nature* **481**, 488 (2012).
Weninger et al., Phys. Rev. Lett. **111**, 233902 (2013).
Weninger and Rohringer, Phys. Rev. A **90**, 063828 (2014).

Reaching even shorter wavelength – Copper K-a lasing at 1.4 Å

Lasing in the hard x-ray regime:

+ higher fluorescence than Auger branching ratio
+ larger oscillator strength and higher gain
- shorter sub-fs core-hole lifetimes
- higher pump-intensities required > 10^{20} W/cm^2

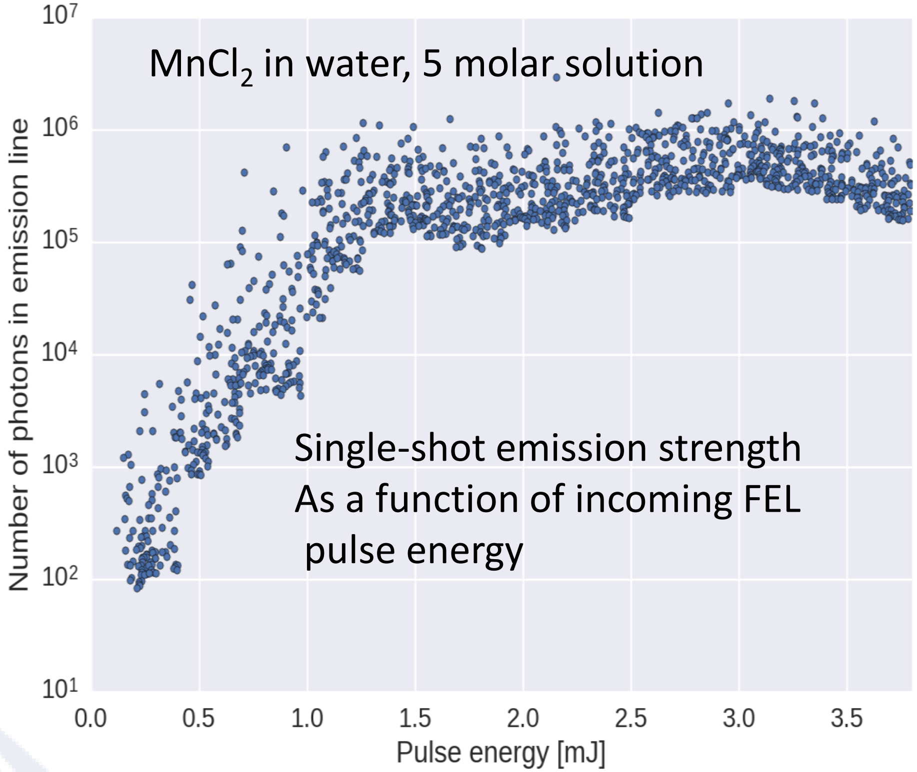
Cu 8,04 keV (solid)

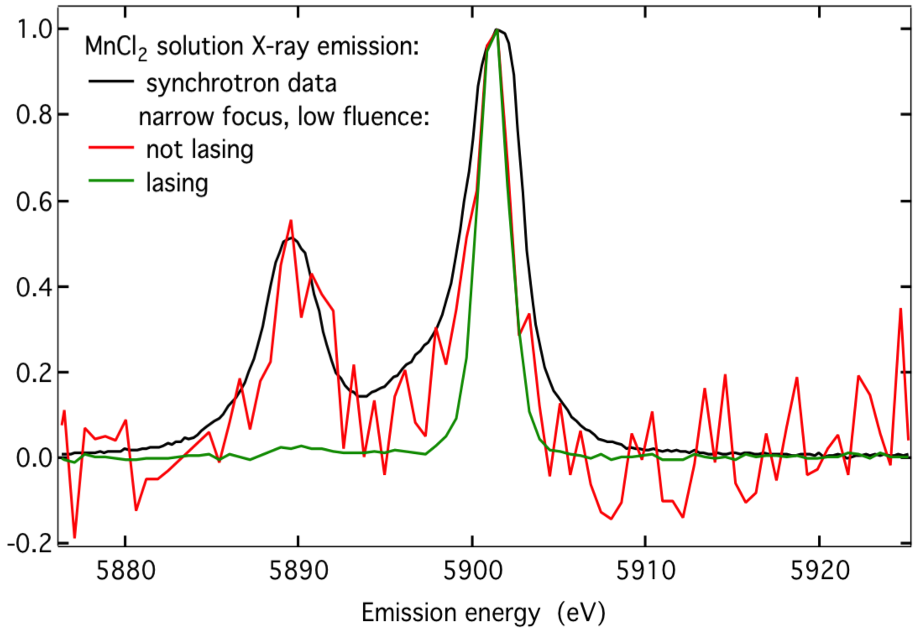


Yoneda et al.,
Nature **524**, 446 (2015).

Hard x-ray laser,
seeded by 2-color FEL operation

Mn K-α x-ray laser for chemical analysis

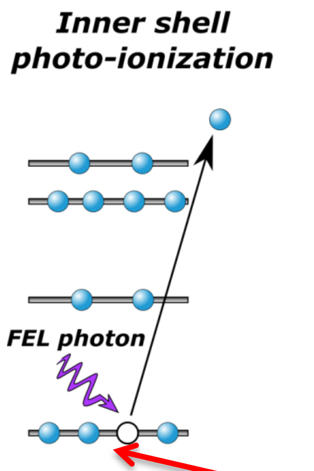




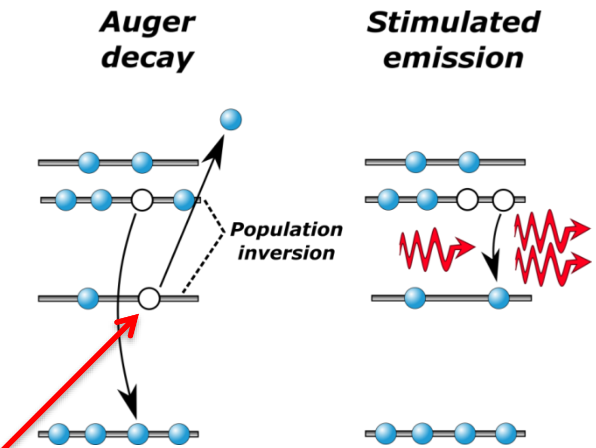
- 1^{st} observation of amplified spontaneous emission in liquids
- Stimulated x-ray emission keeps the chemical information (energy shifts)
- Sharper emission lines (gain narrowing) improves spectroscopic resolution
- High amplification levels allow high-resolution single-shot emission spectra

Slide with courtesy to Th. Kroll

Photoionisation lasing schemes in the XUV domain
Auger-decay pumped XUV laser



Auger decay can lead to
population inversion
in the doubly charged ions



fs Auger lifetime

ps radiative lifetime

Photoionisation lasing schemes
- relatively short Auger lifetime (fs)
- pump intensities $> 10^{15}$ W/cm^2
- small oscillator strength, low gain
- high pulse energies required to achieve saturation

Auger-pumped lasing
- relatively long radiative lifetime (ps)
- pump intensities $> 10^{13}$ W/cm^2
- emission at longer wavelengths

Evidence of stimulated emission
Si surface L edge, emission at 85-100 eV
(gain of factor 2)
M. Beye et al., Nature **501**, 191 (2013)

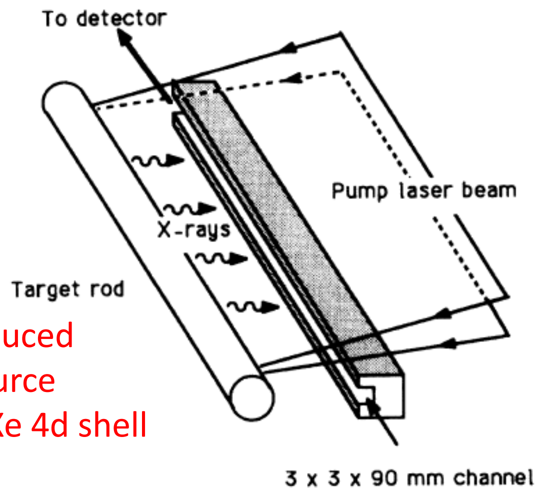
Concept of Auger Laser (Na):
E. J. McGuire, Phys. Rev. Lett. 35, 844 (1975).
1st realisation in Xe and Kr:
H. Kapteyn, R. W. Lee, and R. W. Falcone, PRL **57**,2939 (1986).
H. Kapteyn and R. W. Falcone, PRA **37**, 2033 (1988).

Amplified spontaneous emission in Xe following 4d ionization

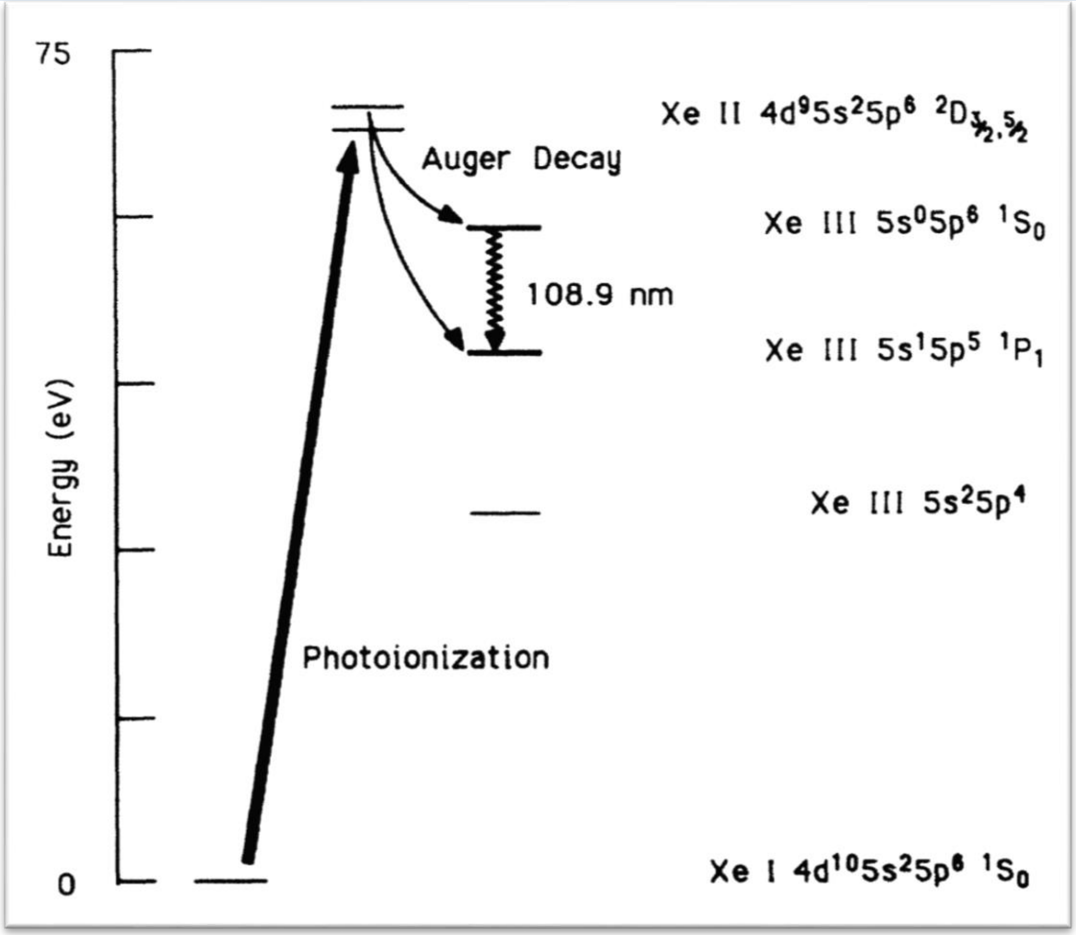
“Giant resonance” for photoionisation of 4d shell at 92 eV

1980s: quest for short wavelength lasers

H. C. Kapteyn, R. W. Lee, and R. W. Falcone Phys. Rev. Lett. 57, 2939 (1986).



Laser-produced plasma source to ionise Xe 4d shell



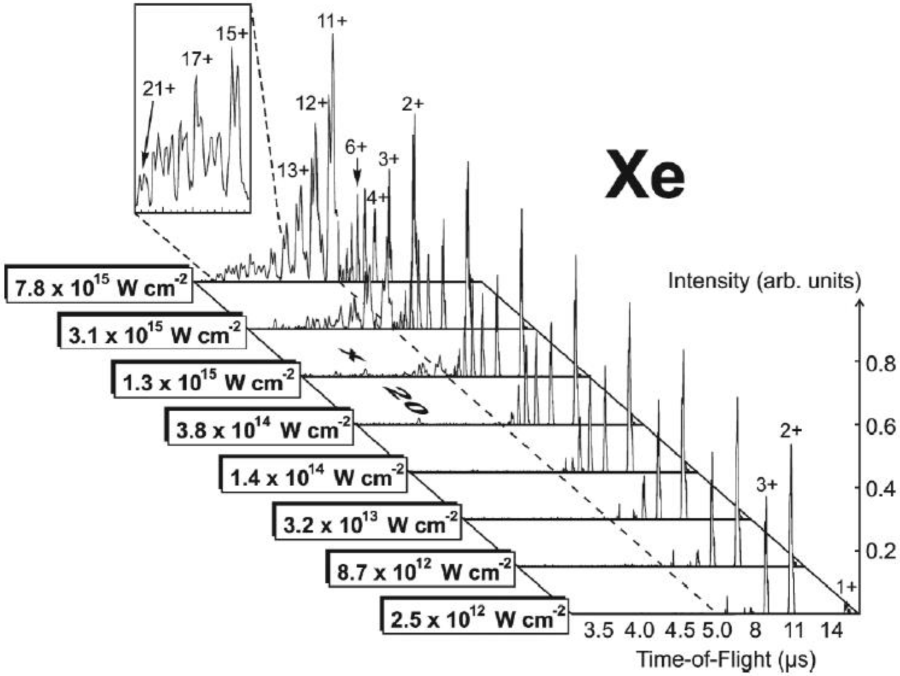
Improved set-up achieved gain of $e^{3.2}$ = 24.5, using 0.56 J @1064 nm on target

G.-Y. Yin, C. P. J. Barty, D. A. king, D. J. Walker, S. E. Harries and J. F. Young, Optics Letters 12,331 (1987).

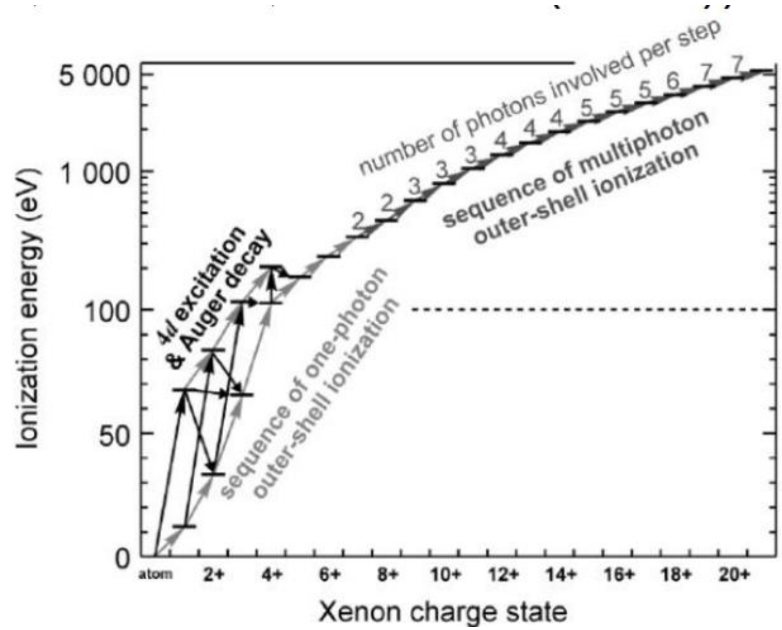
2007: Photoelectric effect at high intensities

Xe = [Kr] $\boldsymbol{4d^{10}\,5s^{2}\,5p^{6}}$

Xe^{21+} observed with I = 7.8×10^{15} W cm^{-2}



A. Sorokin et al., PRL 99 (2007)



Today

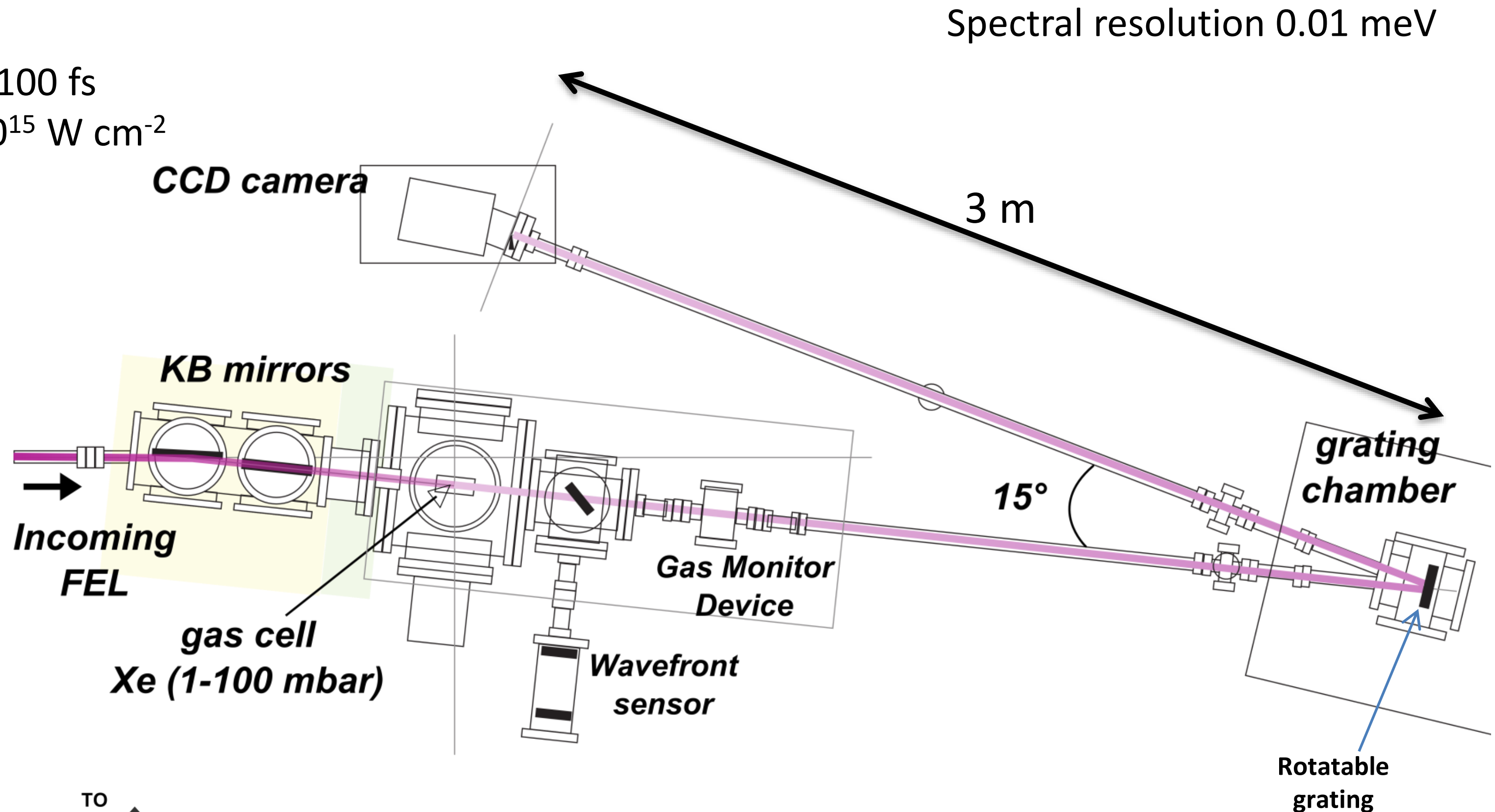
Revisit the schemes of Auger-pumped lasers in Xe with FEL as a photoionizing source

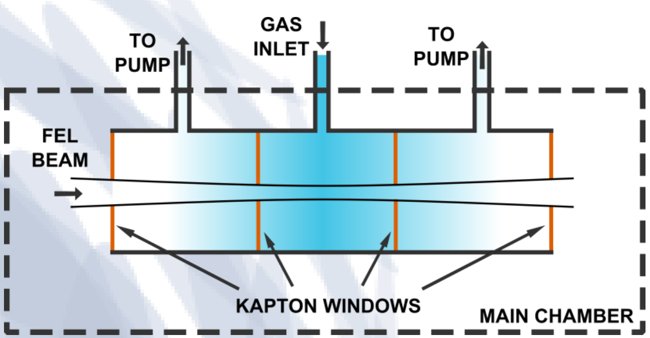
- Can we observe lasing in highly charged Xenon?
- Can lasing in low charge states be quenched by further ionization?
- Can we observe transient soft x-ray lasing in 4d shell?

Experiment at FLASH BL1 beamline

Spectral imaging of focused FEL beam in Xe gas cell

Focus area ≈ 5x6 μm^2
Max energy: 75 µJ
Pulse duration: 50-100 fs
Max intensity: 3×10^{15} W cm^{-2}

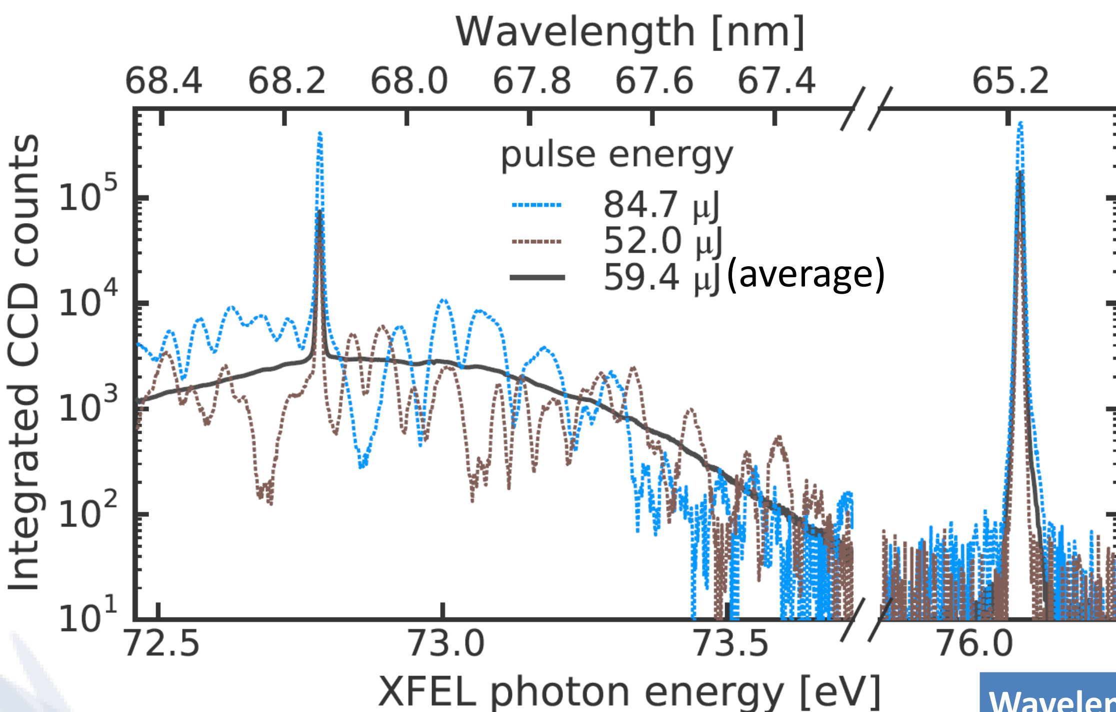




Photon energies investigated: 72 eV and 92 eV

Auger-decay pumped XUV laser

Recorded spectra show FEL radiation in 4th order superimposed to emission lines in 1st order

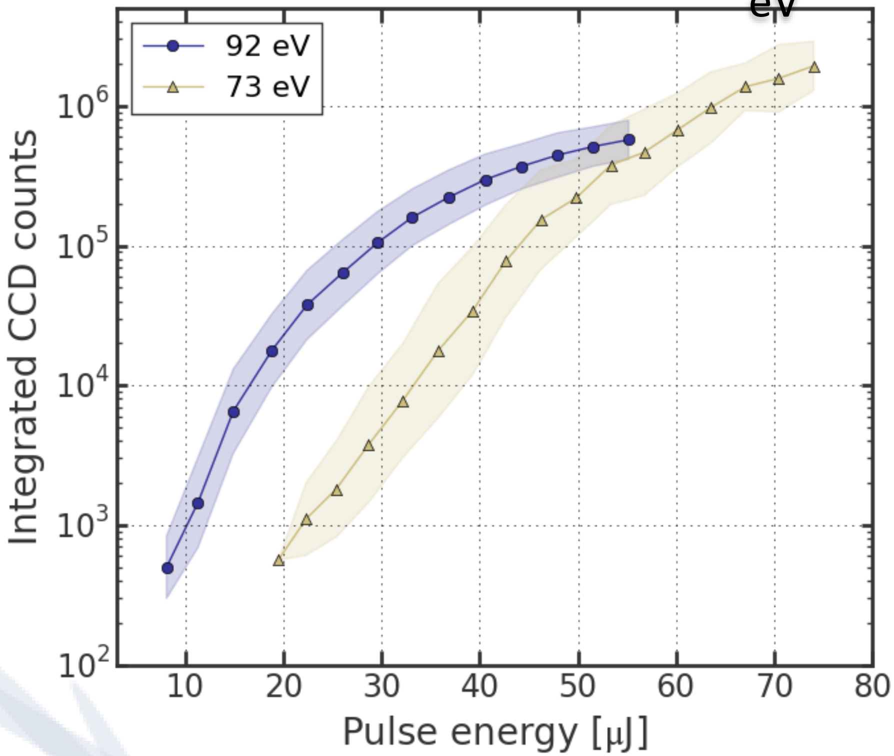


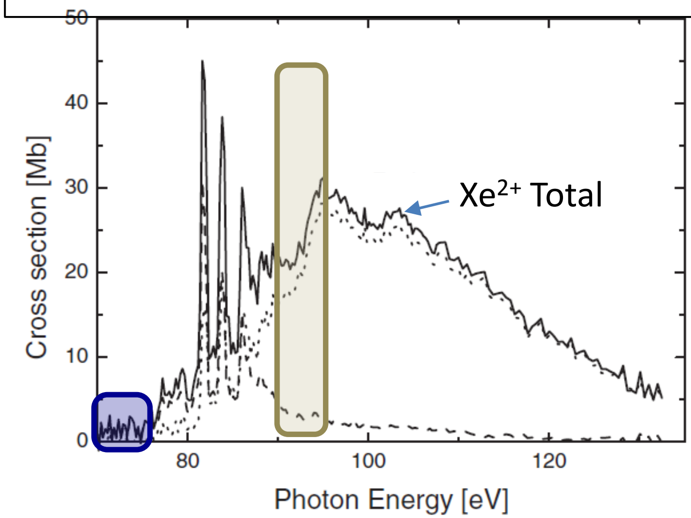
ω_{FEL}= 73 eV

Wavelength [nm]	Energy [eV]	FWHM [meV]	Lifetime [ps]
65.18±0.20	19.02±0.06	1.3	0.5
68.14±0.20	18.20±0.05	1.0	0.7
109.35±0.53	11.34±0.05	0.56	1.2

Gain curves of 65 nm emission line in Xenon

Same lasing lines observed for ω_{FEL} of **72** and **92** eV



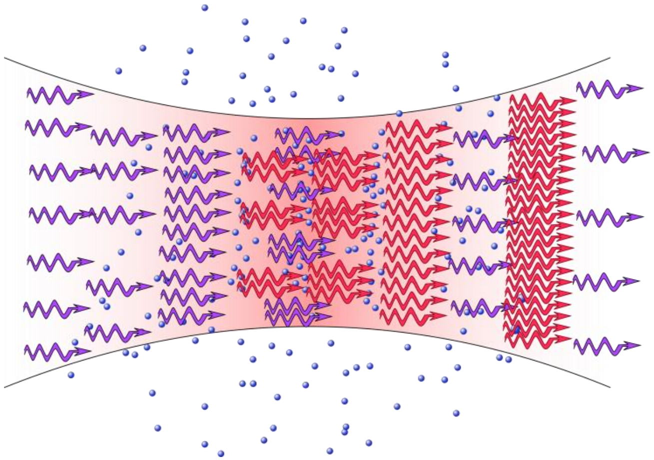


P. Andersen et al., J. Phys. B ***34*** *(2001)*

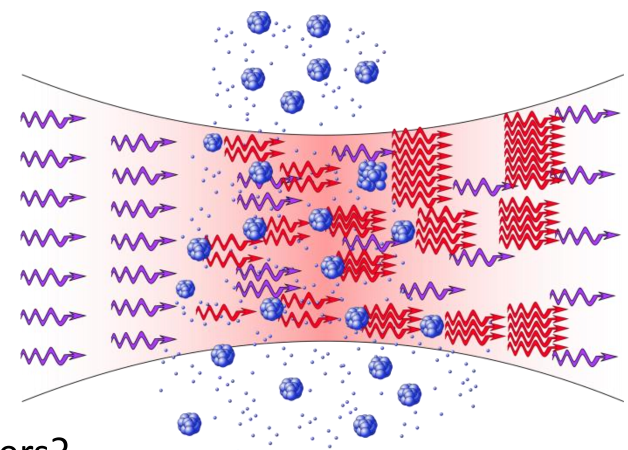
- Exponential growth over 4 orders of magnitude
- Similar gain behavior for emission lines at 68.2 and 108.9 nm
- Due to higher 4d ionisation cross section at 92 eV higher gain at lower pulse energy
- Quenching of lasing at 92 eV due to sequential multiphoton ionisation and strong absorption

From homogeneous medium to quantum emitters
Lasing in clusters

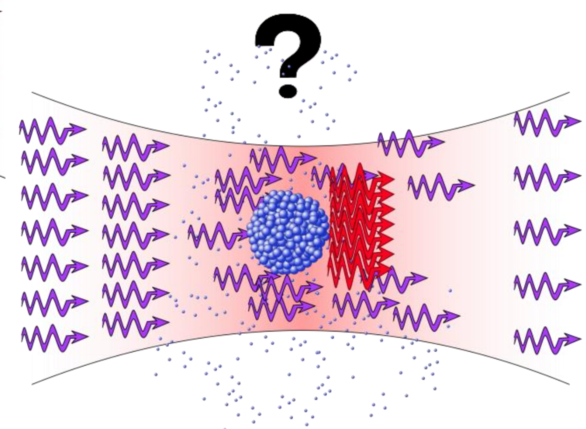
From homogeneous gas…



…to small clusters…



…to single big cluster



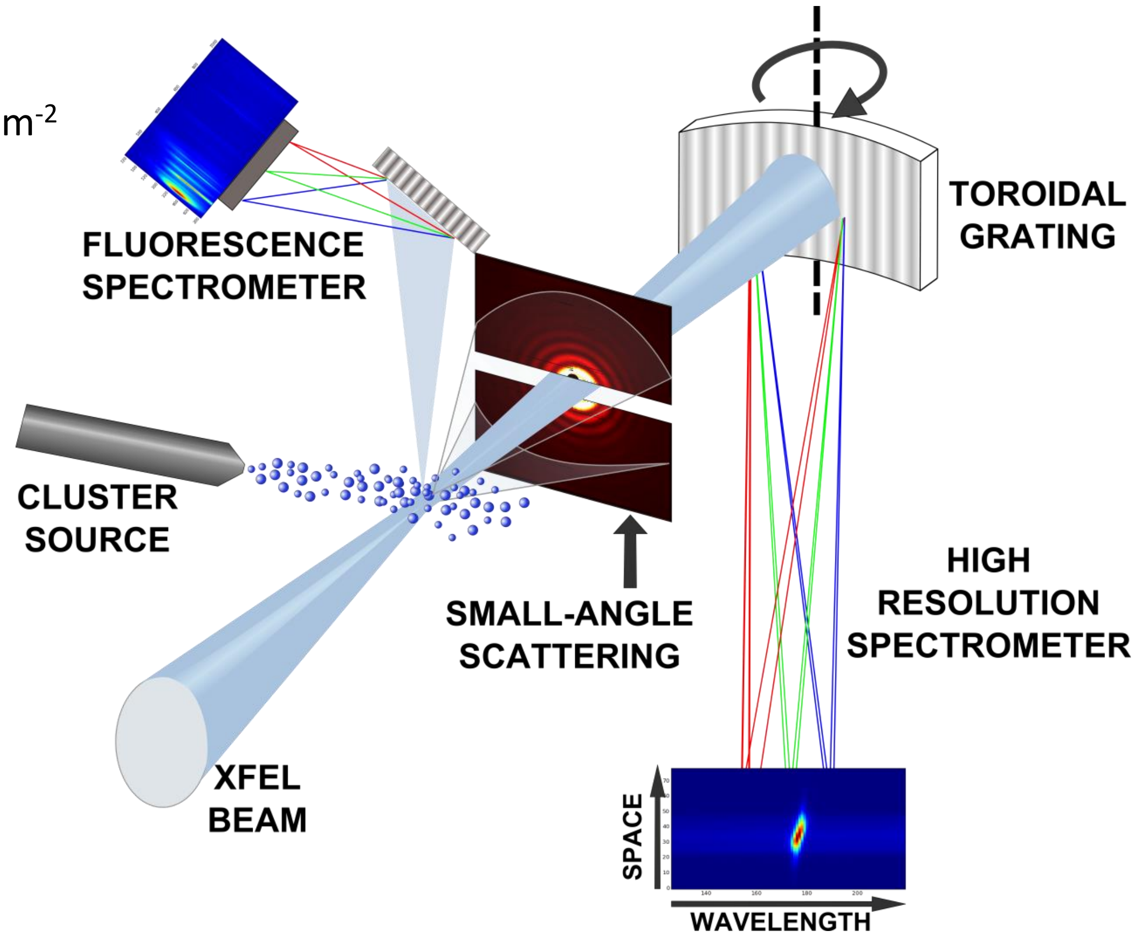
FEL photons
Laser photons

- Can we observe lasing from clusters?
- Are the nano-emitters coherently adding up to the lasing signal?
- Is light amplified and scattered from nanoparticle to nanoparticle?
- Lasing from a single, big cluster?

Experimental Setup (FLASH BL1 beamline)

Focus area:5x6 μm^2
Max. pulse energy: 150 μJ
Pulse duration: 50-100 fs
Max. intensity: 6×10^{15} W cm^{-2}

Spectral resolution 0.01 meV

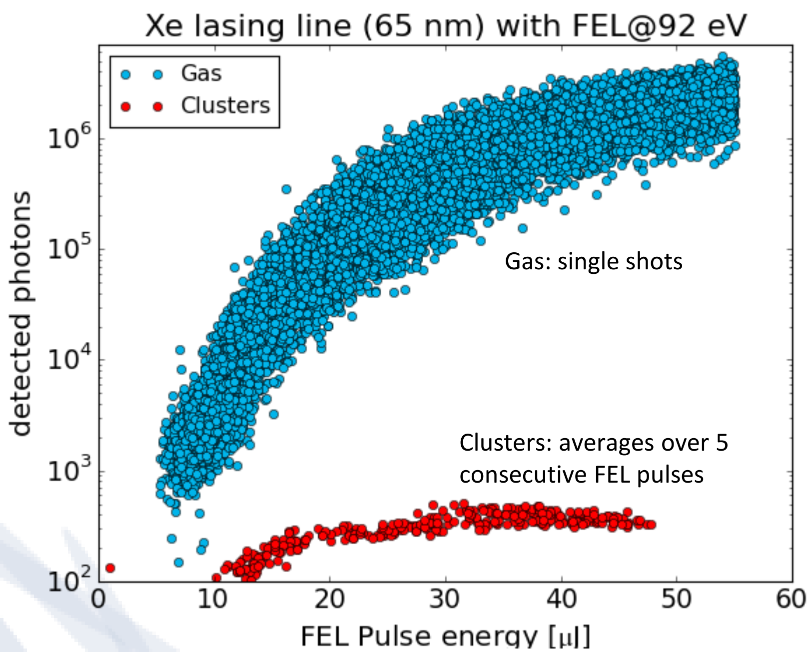


Photon energies investigated: 72 eV, 92 eV, 136 eV

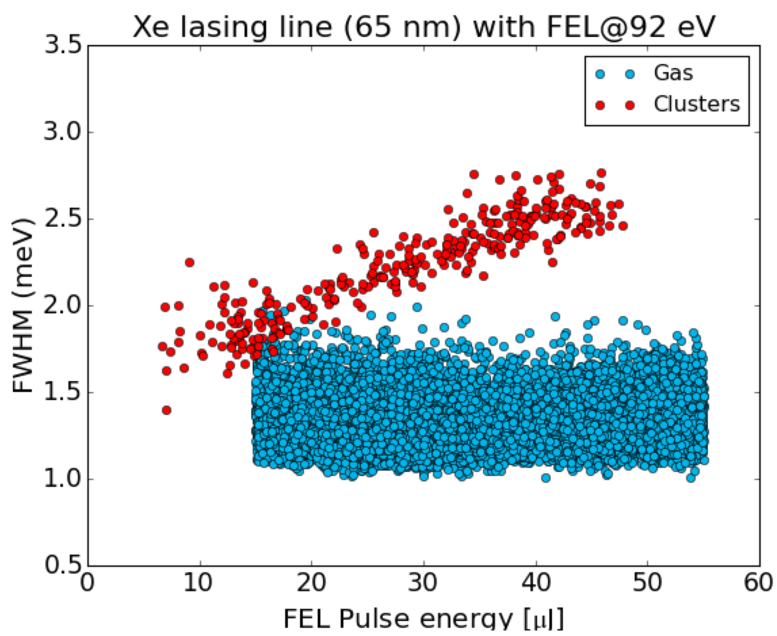
Lasing in clusters versus in gas

Gas: 10 mbar
Clusters: p=5 bar (gas pressure at the nozzle)
T=195 K

Lasing yield



Emission-line width

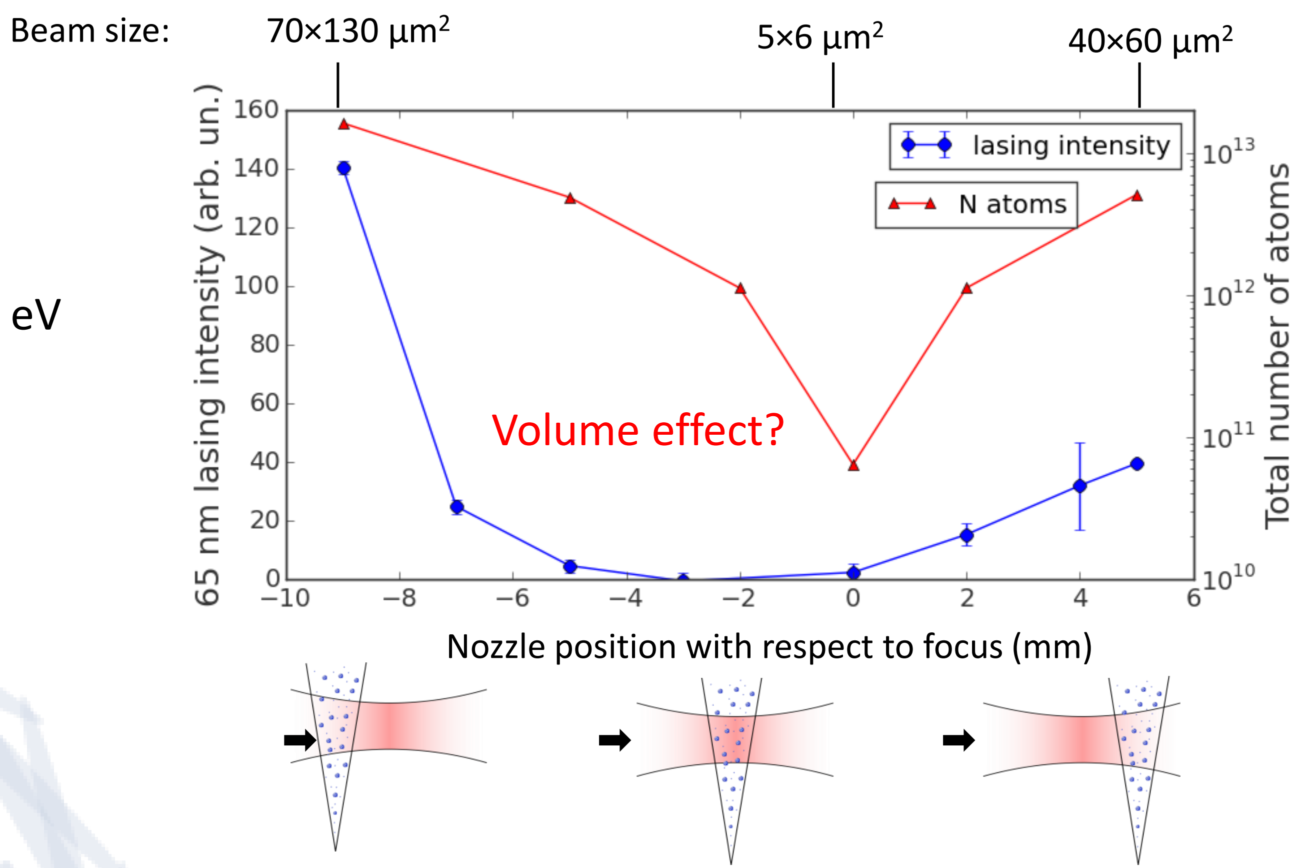


- Saturation in clusters (smaller optical density compared to gas)
- Line broadening due to ionization gating (collisions) for higher pump-pulse energies
- Inverse line width ranges from 180 - 440 fs, compared to pulse duration of 50-100 fs

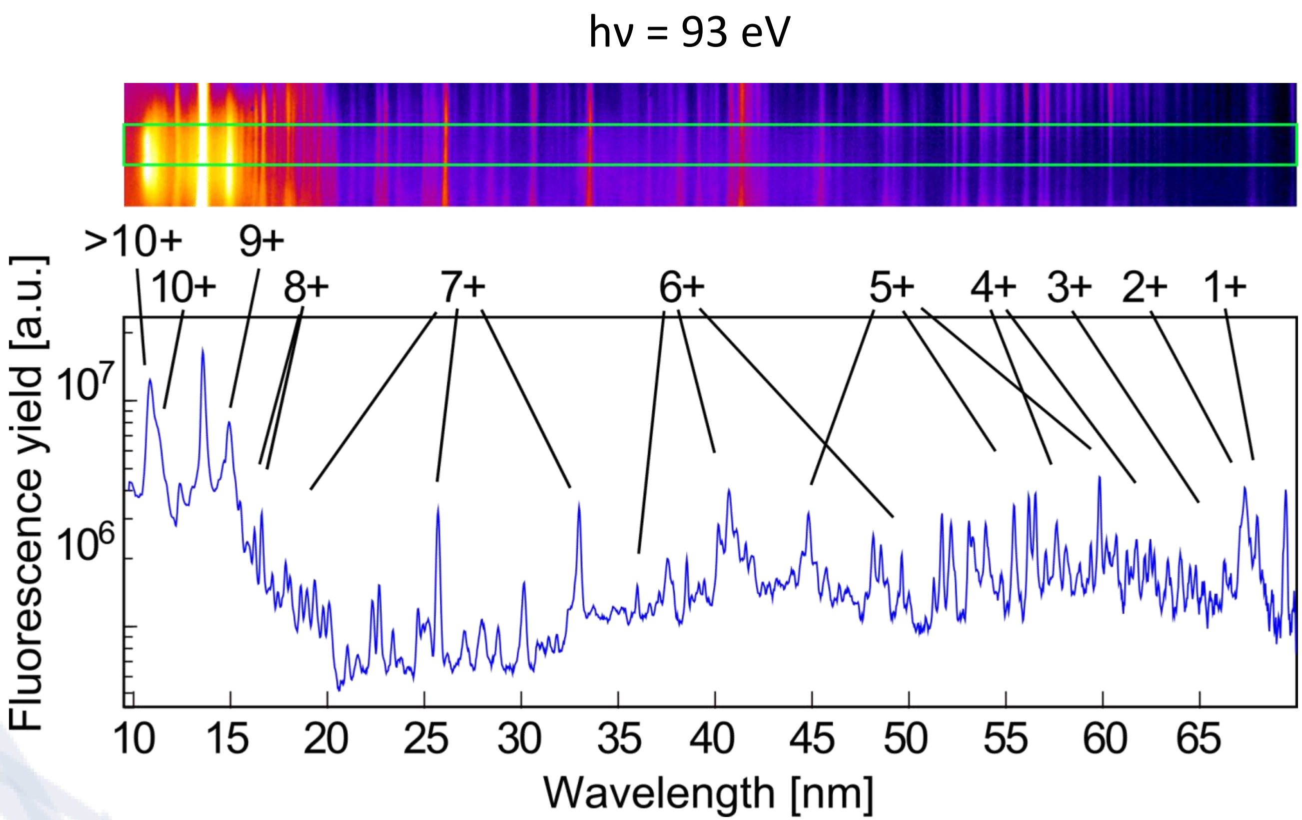
Quenching of lasing at high intensities

Dependence of the emission yield (of the 65 nm line)as a function of pump intensity, keeping the total pulse energy constant

ω_{FEL} = 92 eV



Fluorescence spectrum measured transversely to the beam propagation



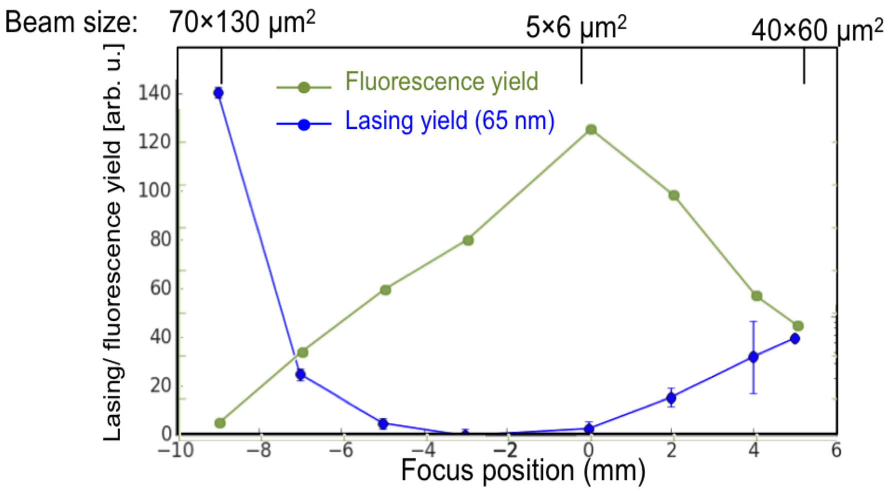
Plot with courtesy to T. Laarmann and A. Przystawik

A nice complementary tool to understand the physics of cluster emission

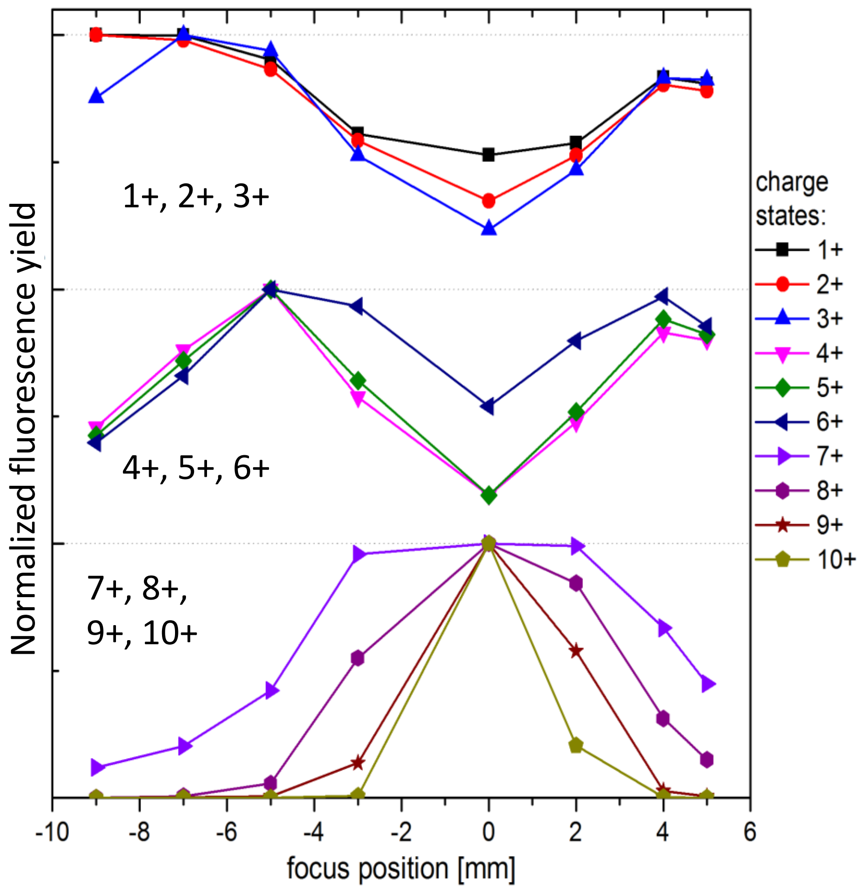
Lasing yield compared to spectrally resolved fluorescence yield

mpsd

Total Fluorescence and lasing yield



Evolution of charge-states



At the focus we observe:

- Drop of lasing signal
- Larger fluorescence yield
- Depletion of low-charge states
- Strong increase of high-charge states

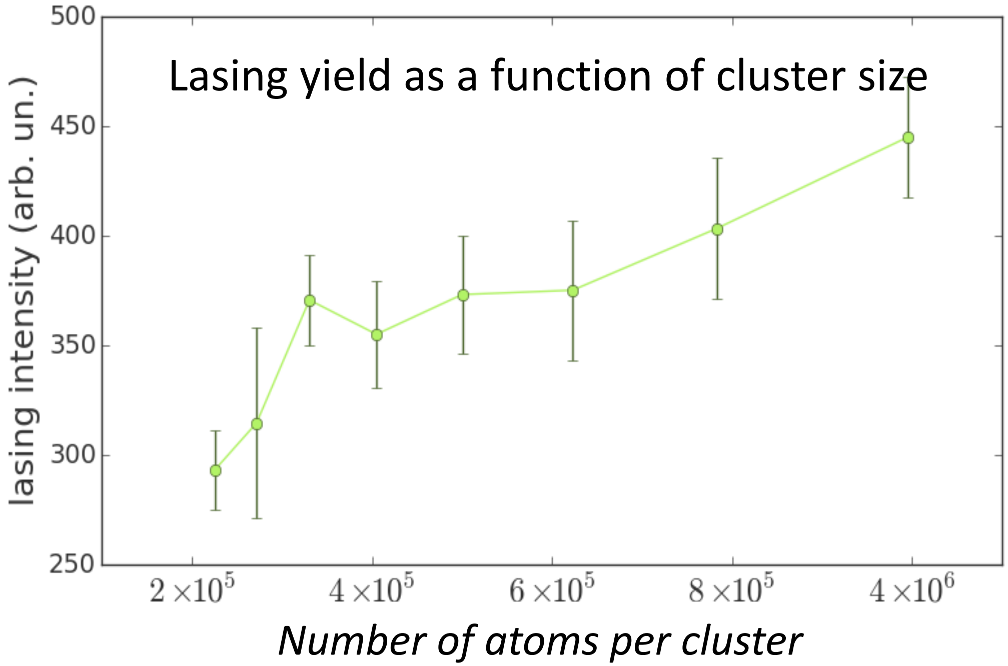
→ Clear observation of quenching of lasing along with creation of higher charge states

→ Quenching of lasing is also observed out of focus by increasing the pulse energy

Lasing in ensemble of clusters as a function of cluster size

Assuming Hagena's scaling laws:
By changing the temperature of the source, the mean cluster size can be changed, keeping the total amount of atoms constant.

$h\nu_{FEL}$ = 93 eV
Lasing line at 65 nm



Total number of atoms is kept constant

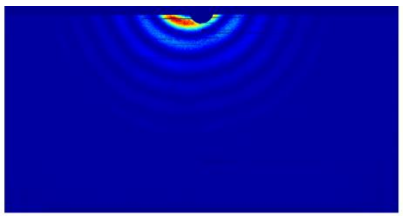
- emission yield grows nearly linearly with the cluster size
- evidence that coherence volume scales with cluster size

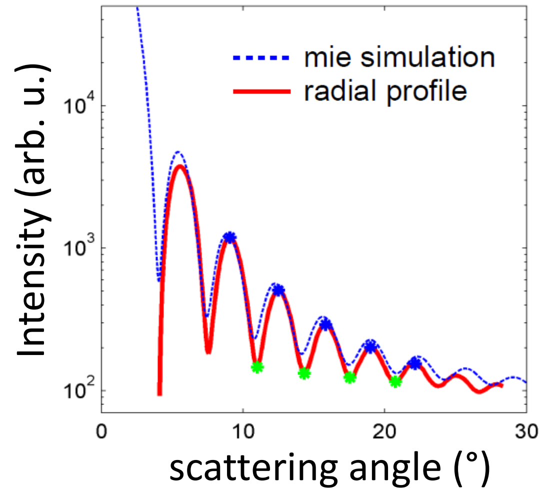
Reaching the limit of a single Cluster

- Single, large clusters can be found in the temporal tails of the pulsed valve emission [D. Rupp et al., J. Chem. Phys **141** (2014).]

- Detect transverse fluorescence signal, lasing signal and scattering image in a single shot

Scattering image



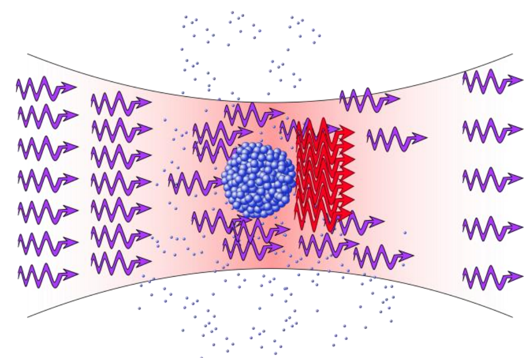


$R \approx 115$ nm ➡ $N_{atoms} = 10^8$

$R \approx 500$ nm ➡ $N_{atoms} = 8\times10^9$

Gas experiment: $N_{atoms} \approx 10^{10}$ - 10^{11}

So far, no lasing signal from a single cluster…



Upcoming experiment with improved cluster set up to chase very large clusters

Conclusions

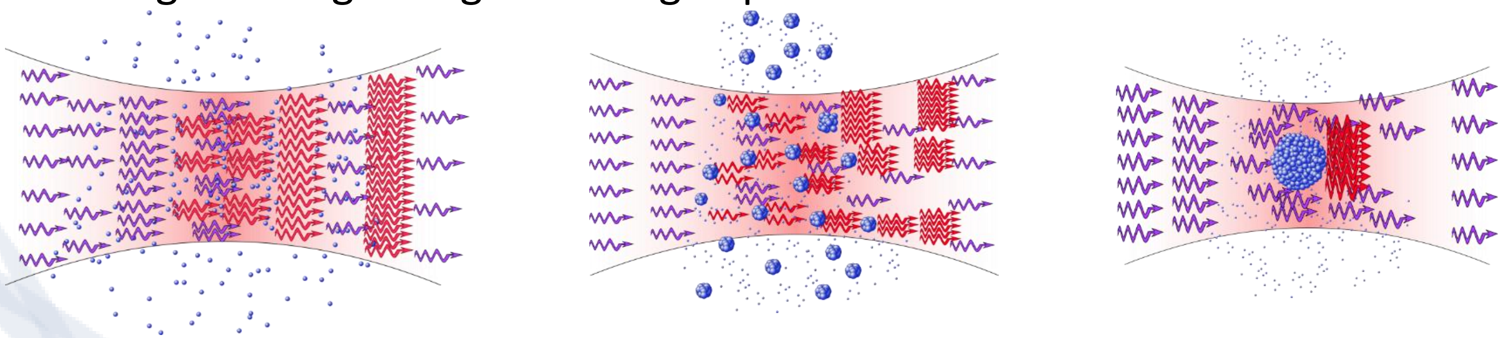
XFELs open the door to study x-ray and XUV stimulated emission

Photoionisation x-ray lasing scheme

- 1st experimental realisation in Neon gas at 1.45 nm
- Demonstration of stimulated K-α emission in Mn salts in solution
    - Gain narrowing leads to enhanced spectroscopic sensitivity
    - Chemical shifts are preserved in strong stimulated emission

Auger-pumped XUV lasing schemes revisited

From homogeneous gas targets to single quantum emitters:

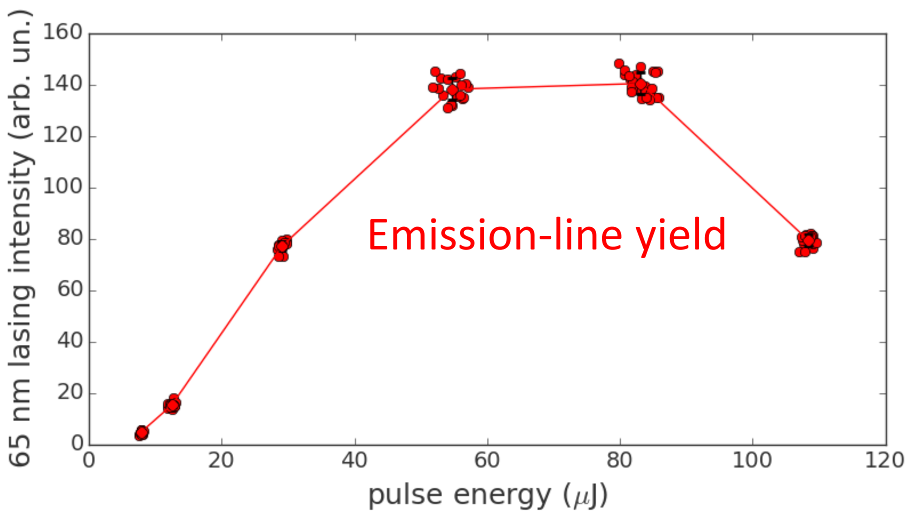


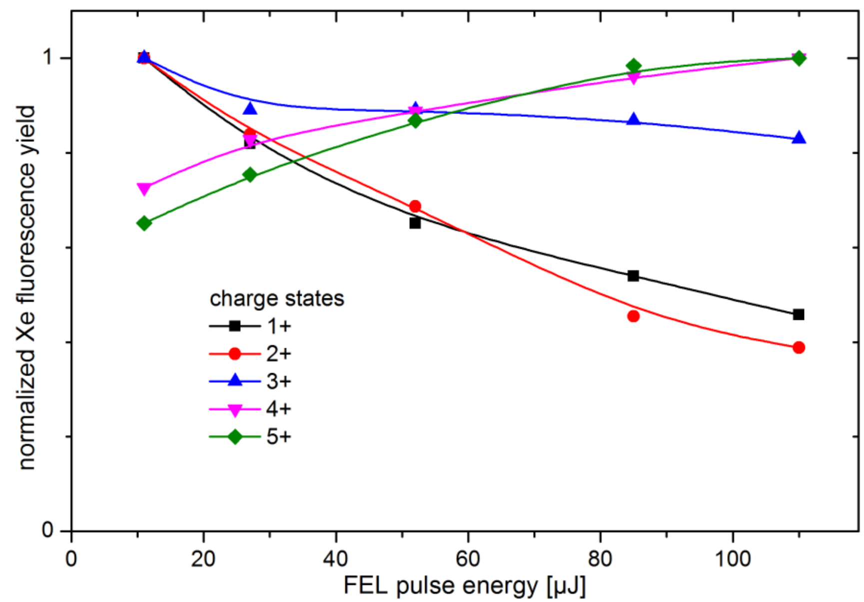
- Study coherent emission of a single macroscopic quantum emitter (cluster)
- Study amplification of ensemble of quantum emitters (random lasers in the XUV)

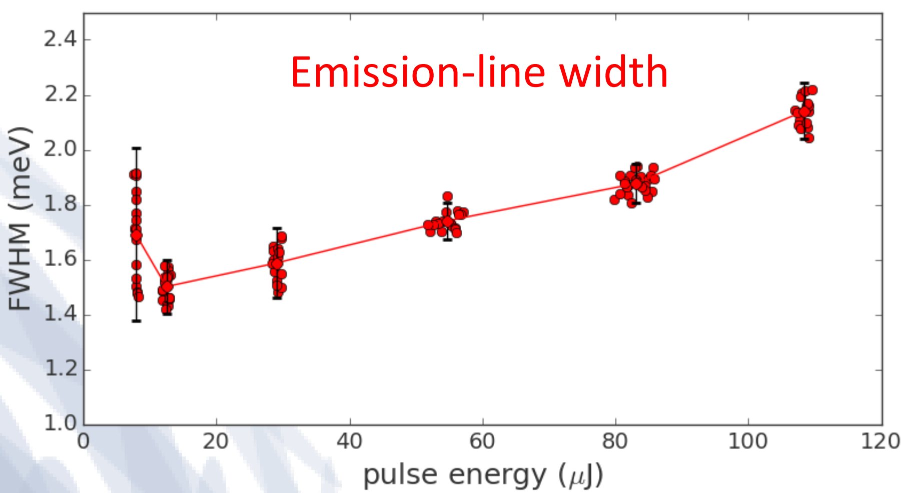
Quenching of lasing is also observed by increasing pulse energy

mpsd

Position of nozzle: 9 mm upstream of focus





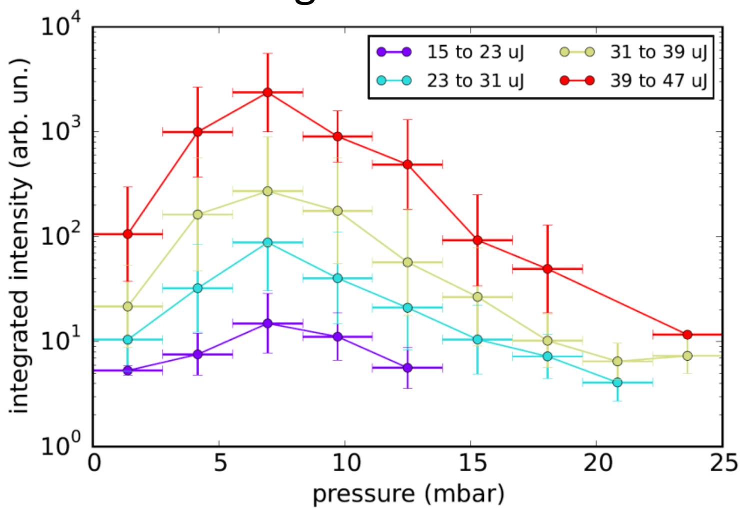


Drop of lasing signal by depleting the lasing states by further collisional ionisation in the cluster.

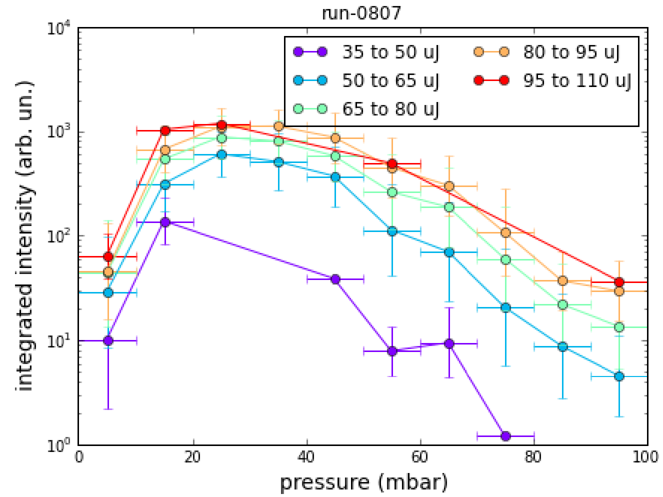
Line broadening and drop of lasing signal was not observed in the gas phase.

Pressure dependence

Lasing at 65.3 nm



Lasing at 108.9 nm



Existence of a pressure optimum for lasing

Pressure optimum different for the two lines

- Transmission at 65 nm ≈ 10%
- Transmission at 109 nm ≈ 100%

Non-homogeneity of the target

Identification of lasing transitions in Xenon

Auger spectroscopy: Mapping electron energies to final upper lasing states

Yates et al., PRA **31** 1529 (1985)

Jauhiainen et al., *J. Phys B **28** 3831 (1995)*

Lablanquie et al., JPB **35** 3265 (2002)

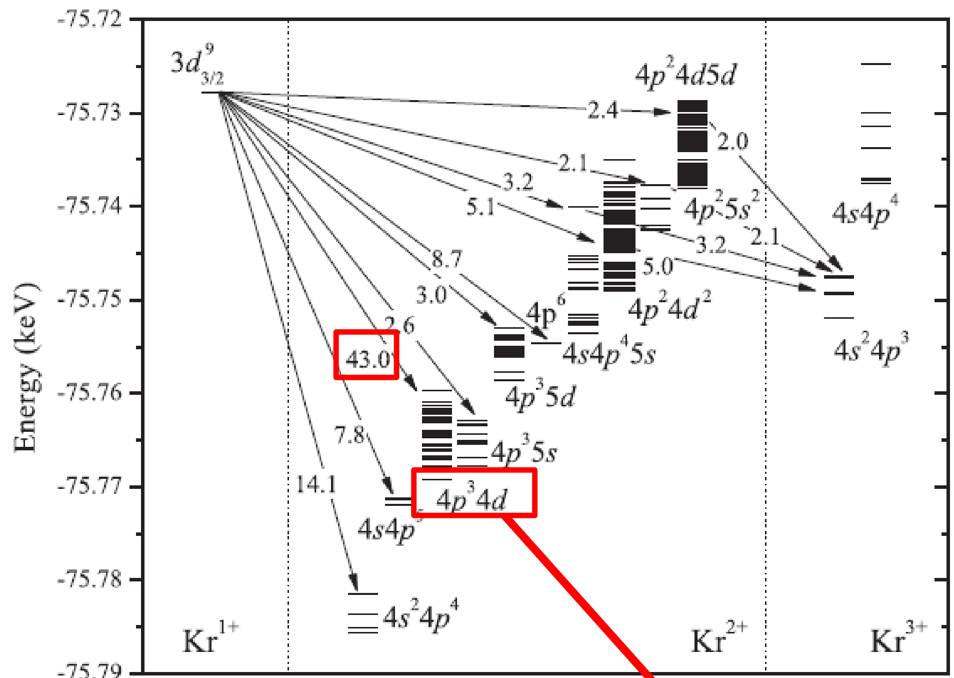
- Strong configurations mixing
- Possible redistribution of states by collisions / valence ionization
- 92 eV opens up new decay channels
- Different branching ratios between 72 eV and 92 eV

Initial State	Ionic state	Energy eV/ purity	Final State	Energy eV / purity	Wavelength (nm)
$5s^0 5p^6$ ($^1S_0$)	Xe^{2+}	26.13 eV 56%	$5s^2 5p^3$ (^2D)5d ($^1P_1$)	14.75eV 44%	108.8954
$5s^1 5p^5$ ($^1P_1$) + 33% $5s^2 5p^3(^2D)5d^1(^1P_1)$	Xe^{2+}	20.27 27%	$5s^2\ 5p^4\ {}^1D_2$	2.12 eV 86%	68.2926
			$5s^2\ 5p^4\ {}^3P_1$	1,21 eV 98%	65.0479
$5s^2 5p^3(^2D)5d\ {}^3P_1$	Xe^{2+}	19.17 20%	$5s^2\ 5p^4\ {}^3P_0$	1.01eV 79%	68.2563
$5s^2 5p^2(^3P)6s\ {}^4P_{5/2}$	Xe^{3+}		$5s^2 5p^3\ {}^2D^*_{5/2}$		65.3695

Identification of lasing transition in Krypton

Experimental uncertainty:
λ = (54.0 +/- 1.4) nm

Krypton Auger decay

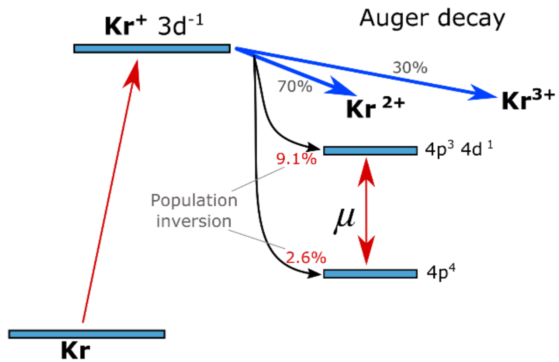


Palaudoux et al., *PRA* ***82***, 043419 (2010)

Observed Wavelength Vac (nm)	Ritz Wavelength Vac (nm)	Rel. Int. (?)	A_{ki} (s^{-1})	Acc.	E_i (cm^{-1})		E_k (cm^{-1})	Lower Level Conf., Term, J	Upper Level Conf., Term, J	Type	TP Ref.	Line Ref.
52.5687	52.5690+	4			0.0	-	190 226.21	$4s^24p^4$ ^3P 2	$4s^24p^3(^2D°)4d$ $^3P°$ 1			L3262
52.8809	52.8811+	2			4 548.4	-	193 651.72	$4s^24p^4$ ^3P 1	$4s^24p^3(^2D°)4d$ $^1D°$ 2			L353
53.0308	53.0309+	4			0.0	-	188 569.14	$4s^24p^4$ ^3P 2	$4s^24p^3(^2D°)4d$ $^3P°$ 2			L353
53.1255	53.1256+	4			0.0	-	188 233.23	$4s^24p^4$ ^3P 2	$4s^24p^3(^2P°)4d$ $^3D°$ 1			L353
53.8544	53.8536+	8			0.0	-	185 688.63	$4s^24p^4$ ^3P 2	$4s^24p^3(^2P°)4d$ $^3D°$ 2			L3262
54.0788	54.0794+	5			5 312.9	-	190 226.21	$4s^24p^4$ ^3P 0	$4s^24p^3(^2D°)4d$ $^3P°$ 1			L3262
54.0860	54.0857+	6			0.0	-	184 891.82	$4s^24p^4$ ^3P 2	$4s^24p^3(^2P°)4d$ $^3D°$ 3			L353
54.3417	54.3417+	5			4 548.4	-	188 569.14	$4s^24p^4$ ^3P 1	$4s^24p^3(^2D°)4d$ $^3P°$ 2			L353
54.4410	54.4411+	4			4 548.4	-	188 233.23	$4s^24p^4$ ^3P 1	$4s^24p^3(^2P°)4d$ $^3D°$ 1			L353

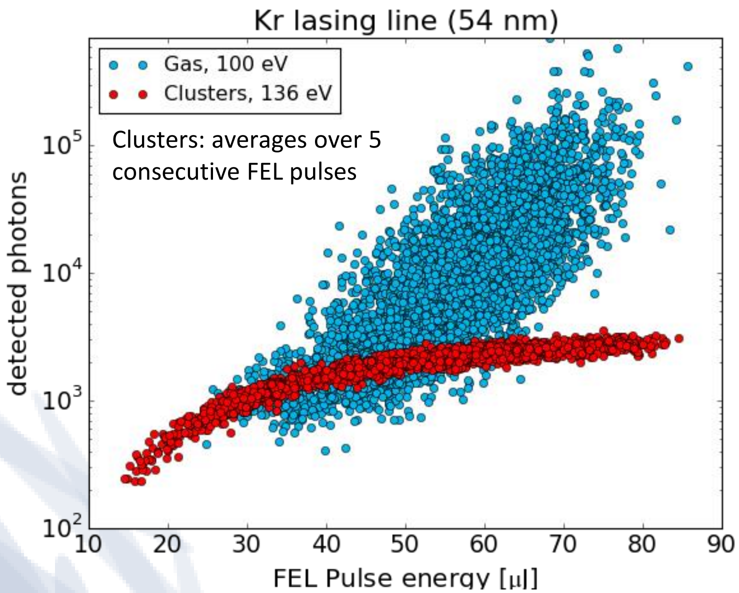
NIST database

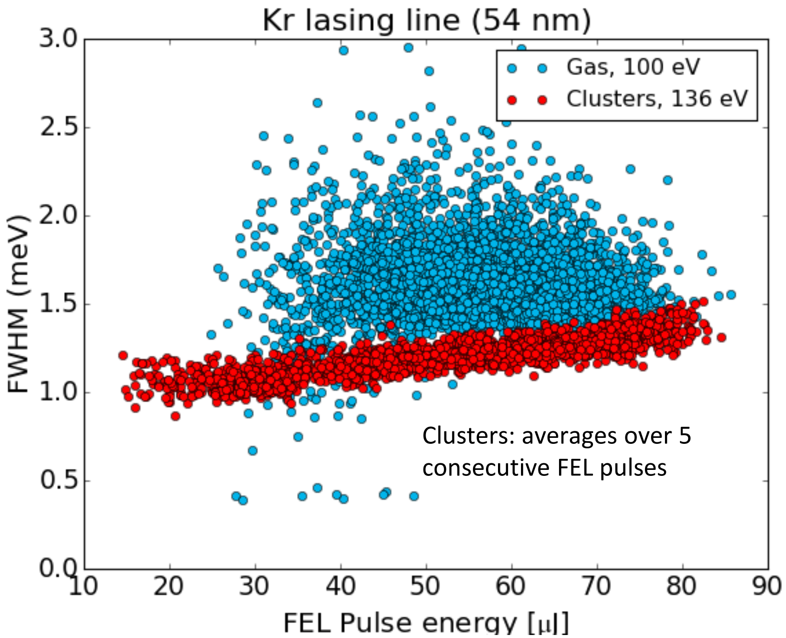
Lasing in gas vs Lasing in clusters (Krypton)



One emission line was detected:
λ=54 ±1.4 nm
$\Delta\lambda$=3.7 pm
ω=22.96 ±0.58 eV
$\Delta\omega$=1.6 meV
Γ^{-1}=400 fs

Gas: 10 mbar
Clusters: P=5 bar (gas pressure at the nozzle)
T=195 K



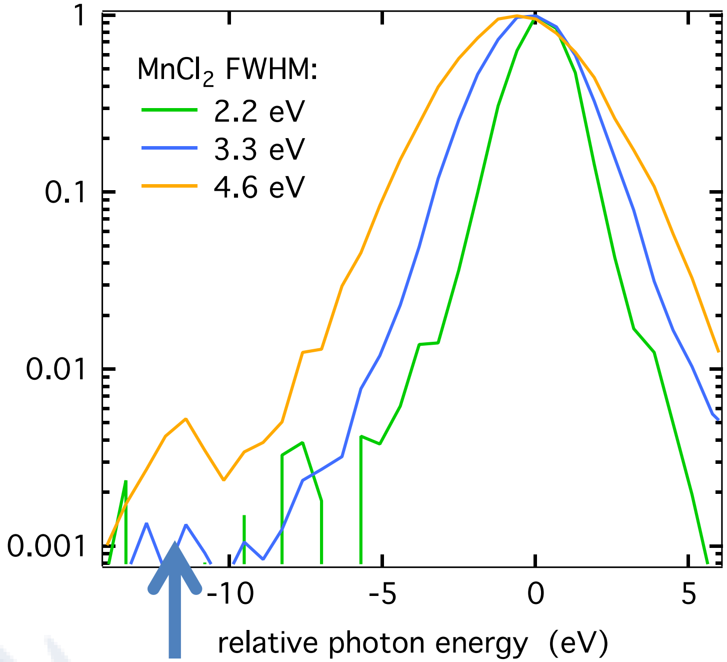


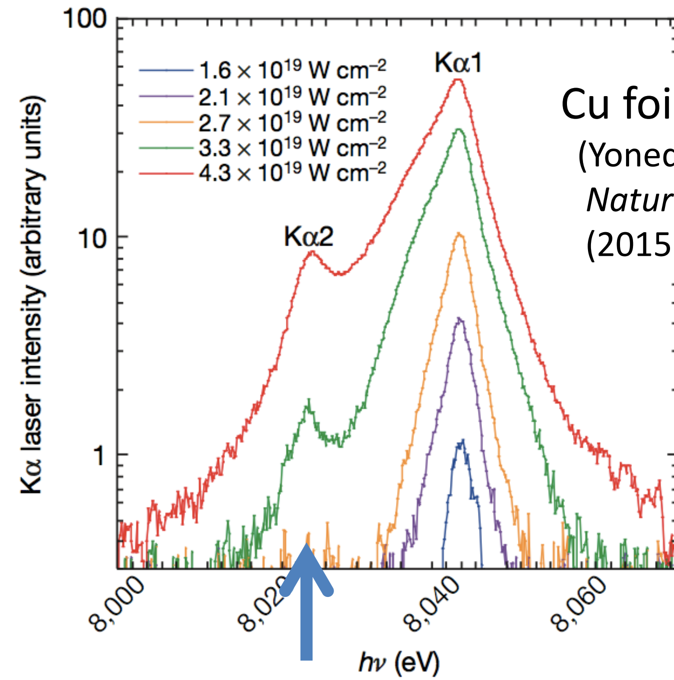
- Saturation in clusters (smaller optical density compared to gas)
- Line broadening due to ionization gating and collisions for higher pump-pulse energies

Shifting and broadening of spectrum at saturation

Emission line profile

Varying pulse energy:





Cu foil
(Yoneda et al., *Nature* **524**, 446 (2015))

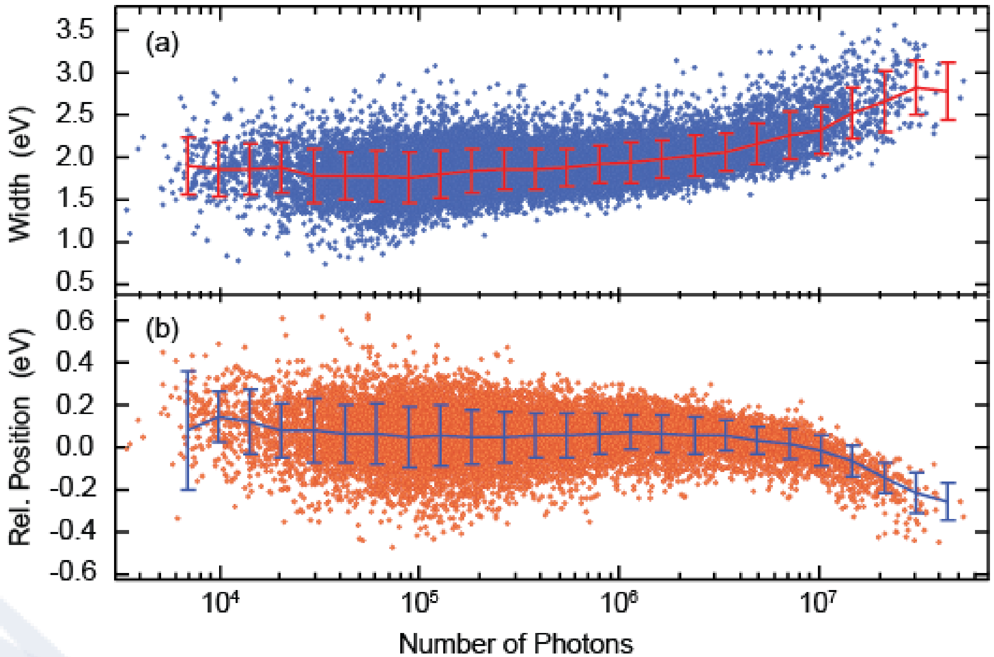
- Increase of $K\alpha_2$ for high fluence (saturation of $K\alpha_1$ transition)
- Broadening and shift due to other multiplet contributions

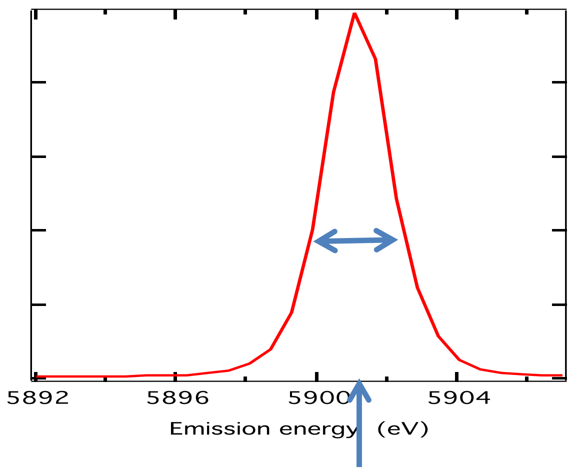
  (compare evolution of spectrum for the molecular x-ray lasers,

  Kimberg and Rohringer, Phys. Rev. Lett. 110, 043901 (2013).)

Line-width dependence of the emission

Peak position and width:





- Low to mid high photon numbers:

  Constant broadening and position

- High photon numbers:

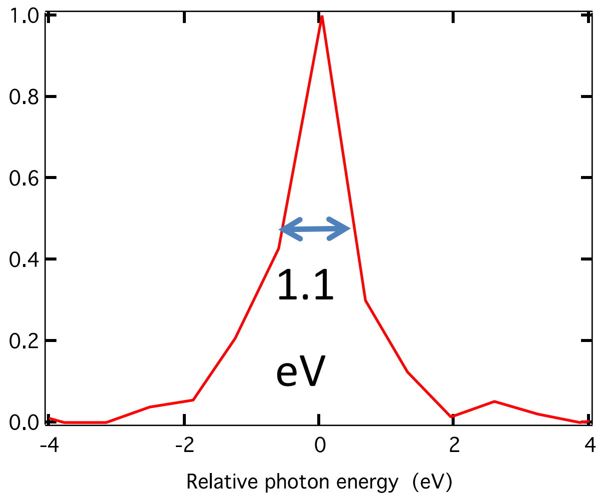
  - Spectral broadening

  - Shift to lower energies

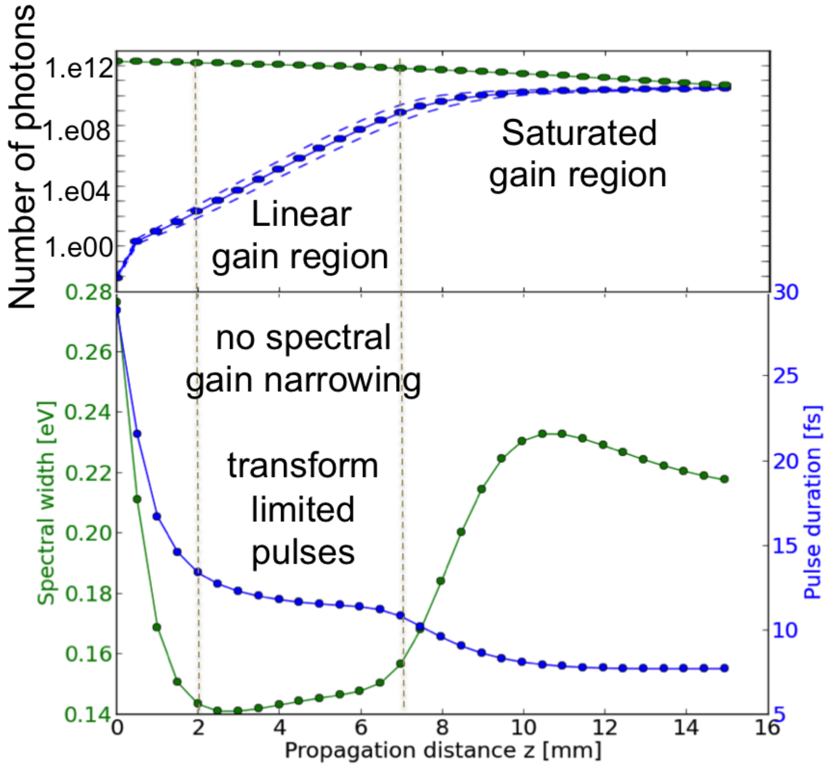
- Variations through beam position, temporal shape, lasing condition

Slide with courtesy to Th. Kroll

Width below life-time broadening at the onset of lasing



- Mn Kα_1 life-time broadening: 1.48 eV (Krause and Oliver, 1979)
- Lowest observed S-XES peak width: < 1.0 eV
- Darwin width of Si (111): 0.77 eV
- Lowest S-XES broadening (gain narrowing): < 0.5 eV



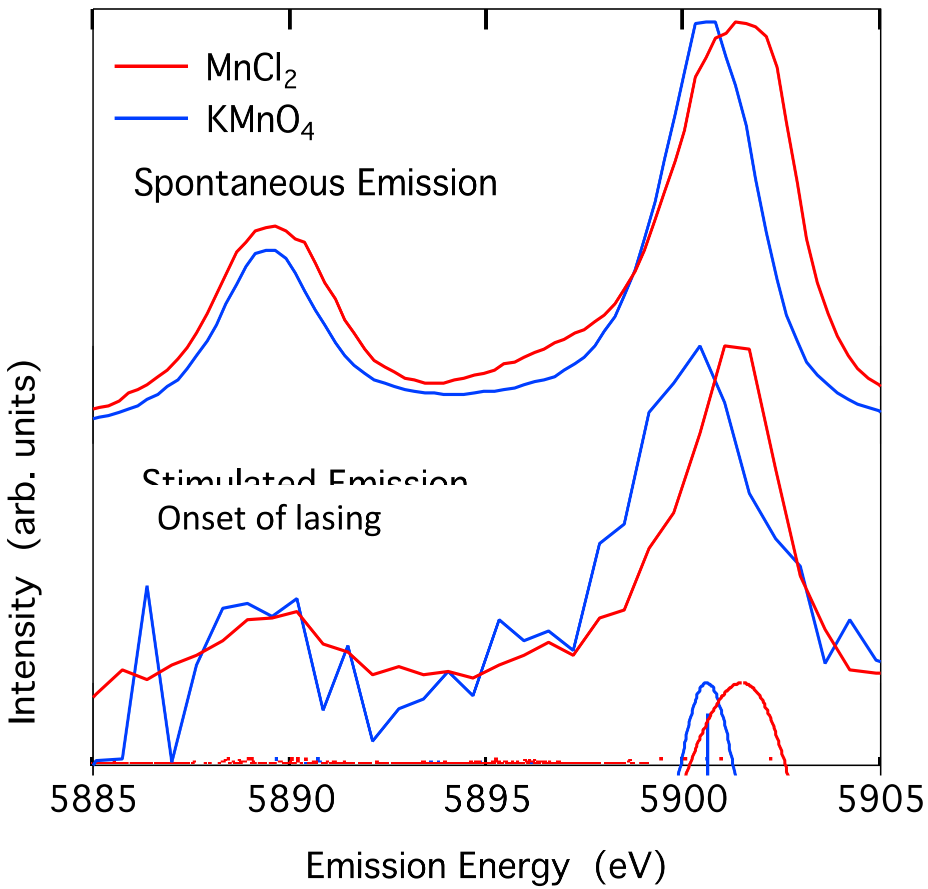
In accordance with predictions of gain narrrowing and saturation broadening (shown for Ne laser)

Weninger and Rohringer, PRA 90, 063828 (2014).

Chemical Sensitivity

Compare solution spectra:

Stimulated emission at the onset of strong lasing retains the expected chemical shift !



Slide with courtesy to Th. Kroll